

REMARKS/ARGUMENTS

Claim 2 has been cancelled without prejudice. Claim 1 has been amended to include the limitations of claim 2; support can be found in original claim 2 and in the specification at page 5, lines 6-9. Support for new claim can be found in the specification at page 19.

No new matter has been added.

Related art rejections

1. The rejection of claims 1, 2, 4, 5, 7, 12 and 14 under 35 USC § 103(a) as being unpatentable over JP 2002-042809 ("*Hara*") in view of US 2003/0054249 ("*Yamamoto*") is respectfully traversed.

The present specification discusses a standard way to produce SiO_x powder, which is to vaporize Si and SiO₂ powders by heating these powders in a vacuum. SiO gas is generated and deposited. The O:Si molar ratio is "almost 1":

SiO powder is produced as follows, for example. A mixture of Si powder and SiO₂ powder is heated in a vacuum to thereby generate an SiO gas, to deposit the gas on a deposition section at a low temperature and to obtain an SiO deposit. A molar ratio of O to Si in the SiO deposit obtained with this method is almost 1.

However, SiO powders produced in this way can further oxidize, thereby increasing the O:Si molar ratio above 1:

The SiO deposit is pulverized to obtain SiO powder and the SiO material is oxidized by oxygen in the air while the SiO deposit is pulverized into powder or SiO powder is used, with the result that a molar ratio of O to Si in an SiO preform exceeds 1. In addition, SiO powder is increasingly more oxidized while the SiO powder is stacked with the powder kneaded coated dried method because of a large surface area of the SiO powder. Therefore, a molar ratio of O to Si is raised in the powder kneaded coated dried layer of SiO.

Therefore, there is little control over the O:Si molar ratio in the SiO film, and the molar ratio can change over time for SiO powders produced in this manner. The result of this additional oxidation is difficulty in releasing lithium ions during battery operation:

With a higher ratio of O to Si in the SiO powder of the powder kneaded coated dried layer, lithium ions occluded in the SiO layer in initial charge are harder to be released during discharge, resulting in a lower initial efficiency.

The above citations are found at page 6 of the specification as filed.

Hara discloses SiO_x powders ("x value is 0.10-1.00") produced in the manner described above: "...the solid state SiO, SiO₂, and Si, or, mixing, being filled up in a reactor and heating at not less than 1400** [presumably °C]." See [0010] of *Hara*. See also [0023] of *Hara*. Thus, one of ordinary skill in the art would recognize that *Hara's* SiO_x films would be susceptible to further oxidation and a lack of control over the oxidation state. This is not remedied by *Yamamoto*, because this cited reference: 1) states that the CVD method is used to "form an amorphous layer with homogenous film quality and an even film thickness"—[0100]; and 2) discloses the use of SiO_x powders where " $0 < x \leq 2.0$ ", where there is no discussion in regard to the criticality of the x value. Thus, there is disclosure in *Yamamoto* that the CVD processes, when applied to *Hara*, would allow for control over the O:Si molar ratio

On the other hand, use of vacuum vapor deposition in the present invention allows for control over O:Si molar ratio, because this ratio can be *intentionally* altered:

A molar ratio of O to Si in silicon oxide forming an negative electrode active material layer of a lithium secondary battery negative electrode of (1-1) is preferably in the range of from 0.5 to 1.2 and especially preferably 0.5 or more and less than 1. That is, in the invention, a molar ratio of O to Si in silicon oxide forming an negative electrode active material layer can be reduced to a value lower than in a case of a powder kneaded coated dried layer. To be concrete, the molar ratio can be not only reduced to a value less than 1, but *intentionally* raised to a value more than 1.

See paragraph bridging pages 15 and 16 of the present specification, emphasis added. This **intentional** control over the O:Si molar ratio in the present invention allows for control over the molar ratio of the SiO anode. This control is supported by the Examples, where the ratio is shown in Table 1, reproduced from US 2007/0059601, in part, below:

TABLE 1

	Film formation material	Average particle diameter (μm)	Negative electrode formation method	Thin film thickness (μm)	Oxygen molar ratio	Initial efficiency (%)	Initial charge capacity	Film formation rate
Example 1	SiO sintered compact	250	Ion plating	0.05	0.5	84	1	
Example 2	SiO sintered compact	250	Ion plating	0.1	0.5	85	1	
Example 3	SiO sintered compact	250	Ion plating	1	0.5	85	1	
Example 4	SiO sintered compact	250	Ion plating	20	0.5	83	1	
Example 5	SiO sintered compact	250	Ion plating	50	0.5	78	0.9	
Example 6	SiO sintered compact	250	Ion plating(*1)	1	0.9	81	1	
Example 7	SiO sintered compact	250	Vacuum vapor deposition	1	1.2	75	1	Slow in resistance heating
Example 8	SiO sintered compact	250	Sputtering	1	1.0	80	1	Extremely slow
Conventional Example 1	—	—	Powder kneaded, coated, dried processing	200	1.4	46	0.6	

There is no suggestion of this feature in either of *Hara* and *Yamamoto*.

In addition, *Hara* discloses that the O:Si molar ratio ranges from 0.1 to 1.00 ("x value is 0.10-1.00"). This range allegedly overlaps with the ranges of the present claims (see claims 4 and 5). However, *Hara* discloses that higher capacities are achieved on the lower end of the x range, and that the capacities **decrease** with more oxygen included in SiO_x:

	SiO _x 量 (g)	フッ酸溶液量 (ml)	SiO _x /HF比	x (SiO _x)	最大放電容量 (mAh/g)
実施例 1	1.0	100	3.0	0.44 ←→	1395
実施例 2	1.5	50	1.7	0.71 ↓	1038
実施例 3	5.0	50	0.5	0.90 ↓	830
比較例 1	---	---	—	1.07 ↓	656

See *Hara's* Table 1, emphasis added.

On the other hand, the present specification states that it is detrimental to have less than 0.5 mole of oxygen (per 1 mole of silicon):

On the other hand, if the molar ratio is less than 0.5, cubic expansion [occurs] when lithium ions are occluded, which causes the negative electrode active material layer to have a possibility of destruction.

See page 16 of the specification as filed, edit added. Thus, the x values of *Hara* that correspond to peak battery performance would be values that destroy the presently claimed batteries. As noted above, *Yamamoto* contains no discussion in regard to the criticality of the O:Si molar ratio. Following the teachings of *Hara* and *Yamamoto* would result in batteries that would fall apart when used for the present invention. Therefore, the present claims would not have been obvious in view of these cited references.

The essence of the *Yamamoto* invention is a combination of the carbon powder negative electrode layer with an auxiliary negative electrode layer of a dense film of silicon oxide formed thereon *to compensate for the small capacity of the carbonaceous negative electrode*. Carbon has a theoretical capacity of 372 mAh/g,¹ while "a theoretical capacity of SiO amounts to several times as large as that of carbon."² In *Yamamoto*, the anode is primarily a carbonaceous material, which is encompassed by a metal oxide: "...when using an anode having a structure according to this invention where an amorphous Sn layer is deposited *on a carbon layer* (curve b)... Then, release of lithium *from carbon* is initiated..." See [0051] of *Yamamoto*, emphasis added. *Yamamoto* implicates carbon as the anode active material:

It is believed that an amorphous layer formed by vapor deposition, CVD or sputtering comprises micropores, through which lithium ions can easily move. Therefore, even when the

¹ [0021] of *Yamamoto*.

² Paragraph bridging pages 2 & 3 of the present specification.

second layer is disposed on the first layer, lithium ions can easily reach a lower carbon material layer, resulting in smooth discharging and charging.

See [0060] of *Yamamoto*, emphasis added. Lastly, *Yamamoto* teaches away from an anode made of metal:

When an anode is made of a metal alone, an electrode potential is increased as in curve d. Thus, in comparison with carbon (curve a), a capacity itself is increased while a range where a battery is used becomes narrower. A capacity cannot be, therefore, increased in a range where a battery is used.

See [0050] of *Yamamoto*.

Accordingly, the structure of the *Yamamoto* anode is a carbonaceous layer encompassed by a metal (oxide) layer. The majority of the anode is the carbonaceous layer:

In this invention, the thicknesses of the first and the second layers may be appropriately determined depending on, e.g., an application. For example, the total thickness of the first layer may be 5 to 1000 μm while the total thickness of the second layer may be 0.1 to 500 μm . ***The total thickness of the second layer is preferably 0.0001 to 0.8 folds both inclusive of the total thickness of the first layer.*** When forming the third layer, the total thickness of the second and the third layers is preferably 0.0001 to 0.85 folds both inclusive of the total thickness of the first layer. Thus, a battery capacity can be increased while maintaining a higher charge-discharge efficiency.

See [0063] of *Yamamoto*. The result is that the *Yamamoto* batteries have capacities close to the theoretical capacity of carbon: 372 mAh/g. See Tables 1 and 2 of *Yamamoto*, where, e.g.

Example 1 has a initial charge capacity of 476 mAh/g.

On the other hand, a dense film of SiO as the anode active material is deposited directly on a collector to a thickness of 0.1-50 μm (see claim 1). Therefore, the majority component of the anode of the presently claimed invention is SiO, which is opposite of what is taught in *Yamamoto*. As a result, the SiO density of the anode is very high and, therefore, batteries using such anodes can reach much higher capacities:

The inventor has had a conception different from conventional in order to achieve the objects, in which it is intended to form a dense layer of SiO on a surface of a collector by vacuum vapor deposition. As a result, it has been found that not only does a capacity per a unit volume in an SiO layer of the invention increases as compared with a conventional SiO layer fabricated with a powder knead coated dried method, but a low initial efficiency, which has been a problem of the conventional SiO layer, is also drastically improved without being accompanied with reduction in initial charge capacity.

See page 5 of the specification.

The above arguments are supported by the Examples, where, as noted in Table 1 above, a battery using a conventional SiO_x powder (Comparative Example 1) exhibits an initial capacity of 60% of the initial capacity of a battery using an inventive SiO_x film in the anode (Example 1). Thus, Applicants have demonstrated that forming an anode by vapor depositing a SiO_x film, where x is defined in claims 4-5 and 20-21, results in batteries having much higher capacities than those of conventional batteries.

There is no suggestion in *Yamamoto* of forming an anode where the majority component of the active material is a metal oxide. Rather, *Yamamoto* discloses hybrid active materials where the carbonaceous substance is the majority component. Thus, applying the teachings of *Yamamoto* to *Hara* would not result in an anode where the active material is a dense, thin film of SiO_x, where the O:Si ratio (*i.e.* the x value) is **controlled** to a value of $0.5 \leq x \leq 1.2$ (claim 4), preferably $0.5 \leq x < 1.0$ (claim 5). Therefore, the invention of the present claims would not have been obvious to one of ordinary skill in the art.

Accordingly, the rejection is no longer tenable and should be withdrawn.

2. The rejection of claims 3 and 13 under 35 USC § 103(a) as being unpatentable over *Hara*, *Yamamoto*, and US 5,755,940 ("*Shindo*") is respectfully traversed for the same reasons given above.

Accordingly, the rejection is no longer tenable and should be withdrawn.

Other matters

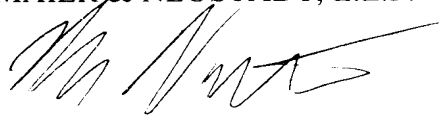
Applicants have complied with the request for a new title. Acknowledgment thereof is respectfully requested.

Conclusion

Applicants respectfully submit that the above-identified application is in condition for allowance. Notification thereof is requested.

Respectfully submitted,

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A handwritten signature in black ink, appearing to read 'Ben Vastine', is written over the printed name.

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